NASA TECHNICAL TRANSLATION

NASA TT F-15,214

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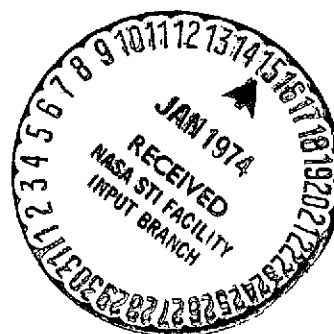
NATIONAL AERONAUTICS AND SPACE ADMINISTRATION
WASHINGTON, D.C. 20546   DECEMBER 1973

1. Report No. NASA TT F-15,214	2. Government Accession No.	3. Recipient's Catalog No.

4. Title and Subtitle SOME ELECTRON STRUCTURE CHARACTERISTICS OF W-Re SOLID SOLUTIONS	5. Report Date December 1973
	6. Performing Organization Code

7. Author(s) S.S. Budagovskiy, V.N. Bykov, M.I. Gavrilyuk, V.N. Pod"yachev	8. Performing Organization Report No.
	10. Work Unit No.

9. Performing Organization Name and Address Leo Kanner Associates, P.O. Box 5187 Redwood City, California 94063	11. Contract or Grant No. NASw-2481
	13. Type of Report and Period Covered Translation

12. Sponsoring Agency Name and Address NATIONAL AERONAUTICS AND SPACE ADMINISTRATION, WASHINGTON, D.C. 20546	14. Sponsoring Agency Code

16. Abstract For solid solutions with a bcc lattice in W-Re alloys, measurement results of various parameters are reviewed. These parameters include the electrical resistance, Hall effect, reluctance, crystal lattice parameter, angular annihilation-photon correlation, and magnetic susceptibility. The possible causes of some of the anomalies exhibited by these data are discussed.

17. Key Words (Selected by Author(s))	18. Distribution Statement Unclassified - Unlimited

19. Security Classif. (of this report) Unclassified	20. Security Classif. (of this page) Unclassified	21. No. of Pages 19	22. Price

SOME ELECTRON STRUCTURE CHARACTERISTICS OF
W-Re SOLID SOLUTIONS

S.S. Budagovskiy, V.N. Bykov, M.I. Gavrilyuk, V.N. Pod"yachev

Re alloying is the most effective means for reducing the /57*
cold shortness temperature of tungsten without worsening the
strength properties at high temperatures [1]. Re has a similar
effect also on two other metals in the VI A group, chromium and
molybdenum. This phenomenon, which is an anomaly within the
framework of the usual concepts on strength and plasticity of
solid solutions, is usually called the Re effect. The heightened
interest in the Re effect was stimulated to a considerable
extent by hopes that understanding the mechanism of the effect
of Re on the cold shortness temperature and the strength of
chromium, molybdenum and tungsten will help to solve a more
general problem, the problem of reducing the brittle rupture
temperature of refractory transition metals.

The existing explanations of the Re effect can be classified
arbitrarily into two groups, extrinsic and natural latticed.
An analysis of various theories is given in the surveys [2, 3],
which imply that the Re effect is apparently caused by a change
in the fundamental characteristics of the solid during the
alloying process. However, the successful development of these
concepts is hindered by the paucity of knowledge about the
nature of the alloys of VI A group elements with Re. This
applies in particular to their electron structure. In this
connection, this article attempts to evaluate, during the /58
alloying process, the character of the change in the parameters
describing the electron structure on the basis of an analysis
of original data and data in the literature dealing with the
properties of W-Re solid solutions.

*Numbers in the margin indicate pagination in the foreign text.

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Galvanomagnetic Properties

The W-Re system alloys (to 25 at.% Re), which in accordance with the constitutional diagram are single-phase solid solutions (Fig. 1), were prepared using the method of electronic zone melting in vacuum without a crucible (EZMWC). EZMWC ensures a high degree of purity, a highly homogeneous Re distribution, a low dislocation density, and a fine structure which is closest to the structure of a completely disordered solid solution. All this makes it possible to study the alloying effect on the electron structure in the purest form, since the transfer phenomena are very sensitive to the structure and alloying elements.
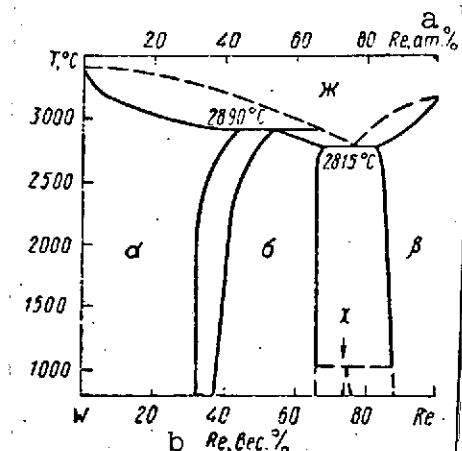

Fig. 1. Constitutional diagram for the W-Re system according to the data in article [4].

Key: a. Re, at.%
     b. Re, wt.%

The original materials used were W and Re powders in which the total content of the admixtures did not exceed 0.02 and 0.05 wt.%, respectively. As a result of EZMWC, the admixture content was reduced, and it did not exceed 0.001 wt.%. In the range up to 6 at.% Re, both mono- and polycrystals of the alloys were used. For higher concentrations, the measurements were made on the polycrystalline samples. The monophase character of the samples was controlled metallographically and by x-ray diffraction methods. In the entire concentration range studied, no precipitation of the σ-phase was discovered. The samples for the measurements were cut out from ingots, using the electrosparking cutting method with subsequent polishing of the surface and etching of the work-hardened layer. To construct the concentration curves, the measured samples were analyzed chemically for Re.

The Hall coefficient and reluctance were measured with direct current on apparatus with a sensitivity of $5 \cdot 10^{-8}$ V. Fields up to 18 kOe were used. The electrical volume resistivity was measured on potentiometer apparatus with a sensitivity of $2 \cdot 10^{-7}$ V. All properties were measured at a temperature of 77°K.

The electrical resistivity curve which depends on the concentration is shown in Fig. 2. The solid lines show the increase in the additional concentration due to the alloying in accordance with the Nordheim rule

$$\rho(x) = Ax(1 - x) \qquad (1)$$

The dependence of the resistivity on the concentration is described well by Eq. (1) up to a 5-6% Re content. For higher concentrations, the resistivity increases faster than as described by Eq. (1), which was extrapolated from low concentrations. For Re contents higher than 8%, the dependence on the concentration can also be described by the Nordheim rule, but with a different parameter A. For an Re concentration above 20%, the values of $\rho(x)$ tend to be smaller than the corresponding values given by the equation. The types of additional resistivities caused by the alloying which depend on the concentrations for alloys of simple metals and simple metals with transition metals were studied in greatest detail in article [5]. The author has shown that for cases when an anomolously large d-state scattering which depends on the alloy concentration is absent, the additional resistivity of the metal is described by the equation

$$\rho(x) = \Sigma A_{\alpha\alpha'} x_\alpha x_{\alpha'}.$$

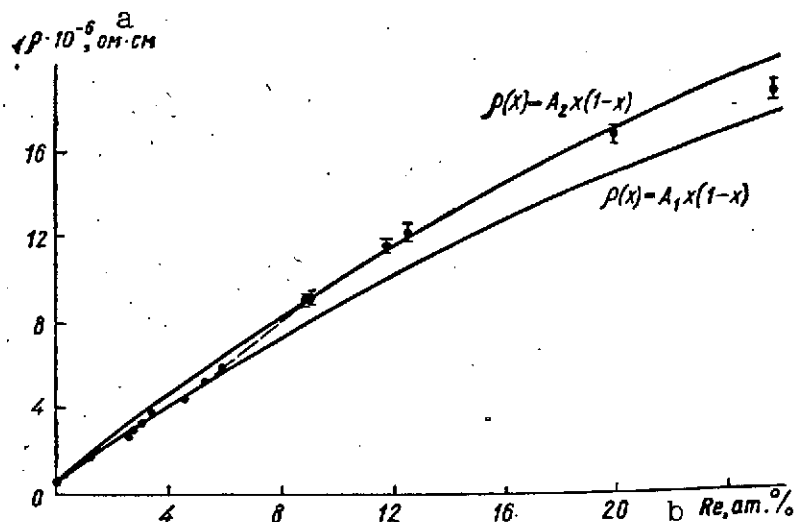For a binary metal, this expression reduces to the Nordheim rule (1).

Fig. 2. Electrical volume resistivity vs. concentration.

Key: a. Ohm·cm
     b. Re, at.%

The applicability of the Nordheim rule to transition metal alloys had not been studied anywhere. It follows from the results of our measurements and from a treatment of the data in article [6] that the change in the volume resistivity in transition metal alloys of neighbor-elements is well described by Eq. (1). The deviation of the values of the electrical volume resistivity from the Nordheim rule in W-Re alloys in which the Re concentrations are higher than 20 at.% is probably related to the deviation of the fine structure of solid solutions from the completely disordered structure resulting from the closeness of the phase boundary (Fig. 1).

It was shown above that the dependence of the additional resistivity on the concentration in solid W-Re solutions, when the Re content is below 5 and above 8 at.%, can be described by Eq. (1) with different values of the parameter A. The parameter A is proportional to the square of the matrix element $|<U_W - U_{Re}>|^2$ where U_W and U_{Re} are the scattering potentials of W and Re atoms respectively [7]; hence, the change in A allows us to assume a change in a certain concentration region in the scattering potential of Re atoms in W.

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The Hall coefficient (Fig. 3) increases sharply without changing signs in the range up to 5-6% Re. For high Re concentrations, the Hall coefficient decreases smoothly and remains positive. For all alloys, the Hall effect depended linearly on the magnitude of the magnetic field, and it was extrapolated at zero when the field tended to zero. The values of the Hall coefficient for monocrystalline samples did not depend on the orientation, and they were the same as the values for polycrystals. This is apparently due to the relatively high temperature and the low effective field.
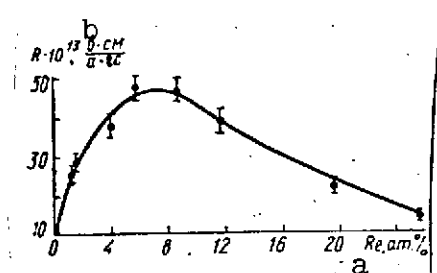

Fig. 3. Hall coefficient vs. concentration.

Key: a. Re, at.%
     b. V·cm/A·gauss

The reluctance could only be measured in tungsten and other tungsten alloys with 1.3 and 1.6% Re, since for higher concentrations, the effect was small. In all cases, $\Delta\rho(H)/\rho$ increased proportionally with H^2.

The positive sign of the Hall coefficient and the reluctance which is different from zero indicate in the W the presence of carriers of both types. Therefore, the use of a two-zone model to describe the properties of W and its diluted alloys is probably admissible. Of course, the two-zone model is only a first approximation, since it takes into account the anisotropy of the Fermi surface. But for the fields and temperature used, anisotropy of the Hall coefficient was not detected, so that this approximation can be used to estimate qualitatively the character of the change in the electron structure during the alloying process. According to the two-zone model [8], the conductivity σ, the Hall coefficient R and the reluctance $\Delta\rho/\rho$ can be written in the form

$$\sigma = \sigma_- + \sigma_+;$$

$$R = \left(\frac{\sigma_-}{\sigma}\right)^2 R_- + \left(\frac{\sigma_+}{\sigma}\right)^2 R_+ ,$$

$$\frac{\Delta\varrho}{\varrho} = \frac{\sigma_-\sigma_+(\sigma_-R_- - \sigma_+R_+)^2 H^2}{(\sigma_- + \sigma_+)^2 + H^2\sigma_-^2\sigma_+^2(R_- + R_+)^2} ,$$

(2)

(3)

(4)

where σ_-, σ_+, R_-, R_+ are the conductivity and the Hall coefficient of the electrons and holes, respectively. Thus, for the fields and temperatures used, the second term in the denominator of expression (4) is four orders of magnitude smaller than the first term, and it can be ignored. We will use the well-known relations $\sigma = evN$, $R = 1/eN$, where e is the charge of the carrier, v is the mobility, and N is the number of carriers per atom. Then Eqs. (2)-(4) take on the form

$$\sigma = ev_+N_+ - ev_-N_-;$$

$$R = \frac{eN_+v_+^2}{\sigma^2} - \frac{eN_-v_-^2}{\sigma^2};$$

$$\frac{\Delta\varrho}{\varrho} = -\frac{e^2v_-v_+N_-N_+(v_- - v_+)^2 H^2}{\sigma^2} .$$

(5)

(6)

(7)

Since for tungsten, $N_- = N_+$ [9], we obtain from Eqs. (5)-(7) the values $N_{-,+}$ and $v_{-,+}$, $N_{-,-} = 0.063$ atom^{-1}. For all the coarseness of the two-zone model, the number of carriers per atom obtained (0.12) has the same order of magnitude as the quantity determined in detailed studies of the Fermi surface [9].

To determine $N_{-,+}$ and $v_{-,+}$ from Eqs. (5)-(7), one of the quantities N_- or N_+ must be used as a parameter. The value of the parameter is selected so that $N_-/N_+ \geq 1$. This criterion was used on the basis of the following considerations.

The basis for the analysis of the change in the zonal structure of metals during the alloying process (it is assumed that the type of crystal lattice does not change) is the

hard zone theorem, according to which the change ΔE in the
energy of the single-electron space in an AB alloy in the first
order perturbation series is equal to

$$\Delta E = < \Psi_k | V_p | \Psi_k >,$$

where Ψ_k is the wave function describing the single-electron
state and V_p is the perturbation potential [10].

In this approximation, ΔE does not depend on the magnitude
of the wave vector k. Therefore, the energy zone is displaced
as a whole without changing its form. The density of the
states in the alloy is defined by the function $N(E) = N_0(E-\Delta E)$;
it has the same form as in a pure metal. Consequently, the
additional electrons introduced by the admixture atoms will
simply fill the zone. The validity of applying this theorem
to the description of the electron structure of transition
metal alloys has been discussed widely [11-13]. An analysis
of a large amount of experimental material [14] allows one to
draw the conclusion that the change in the zonal structure of
transition metal alloys obeys the hard zone theorem, at least
for alloys of neighboring elements (naturally assuming that
the alloying does not change the crystal lattice). In article
[15], the picture for the density of the states for the con-
ductivity zone of tungsten and the energy of the bands was
obtained theoretically (Fig. 4). Using these data, it is
possible to estimate within the framework of the hard band
theory the character of the change in the parameters of the
zonal structure of tungsten as a result of a change in the
number of valent electrons per atom. In particular, during the
alloying of tungsten with rhenium, which probably displays in
alloys with tungsten its high valence [16], the density of the
states on the Fermi surface must increase, and the holes are
filled. But within the range where the solid solutions exist,
the holes will not be filled completely. The results of measure-
ments of the electron heat capacity $C = \gamma T$, where $\gamma \sim N(E_F)$ is
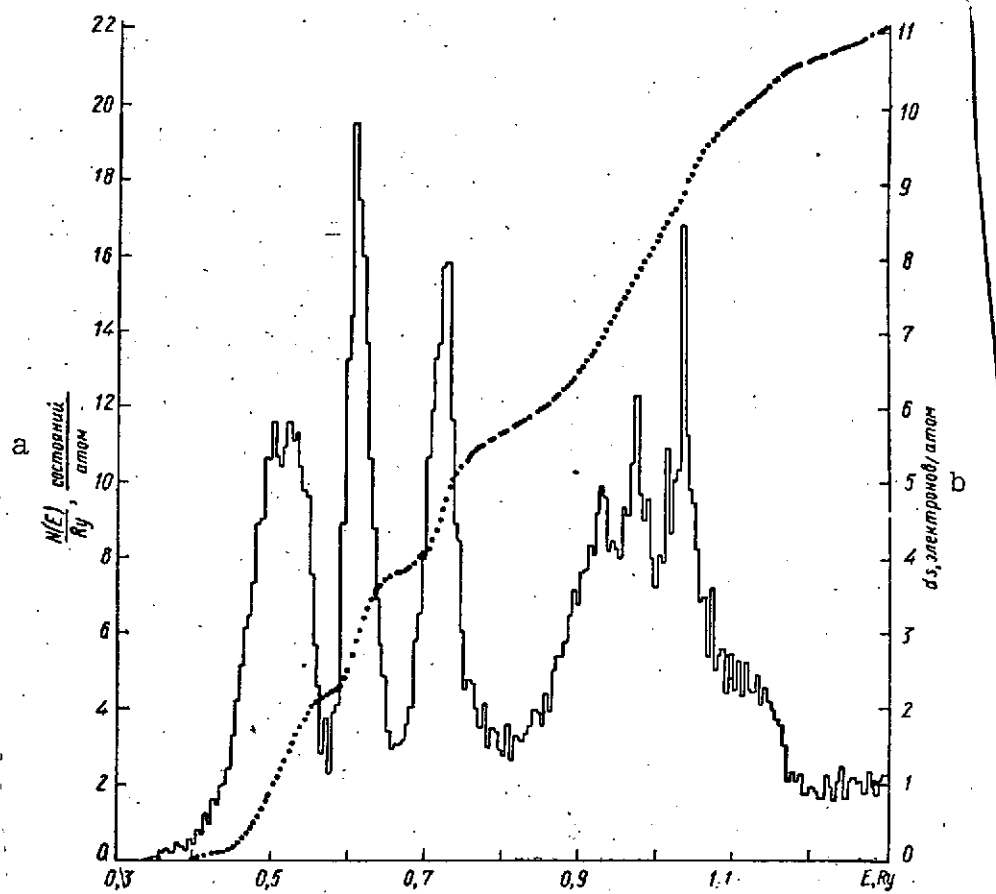
Fig. 4. Density of states in the tungsten conductivity zone [15].

Key: a. States/atom
     b. ds, electrons/atom

the density of the states on the Fermi surface [17] of the alloys under consideration (Fig. 5) apparently allows us to conclude that the changes in the zonal structure of W-Re solid solutions can be described within the framework of the hard band model. The dependence of the temperature on the concentration in the transition to the superconductive state [18] also confirms this conclusion. Therefore, the introduction of the additional condition $N_-/N_+ \geq$ is probably admissible.
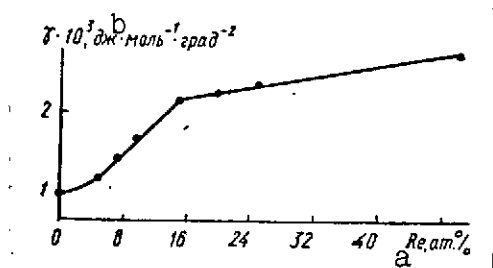
Fig. 5. γ vs. concentration.

Key: a. Re, at.%
     b. J·mole^{-1}·degree^{-2}

The dependence of N_- and N_+ on the concentration (Fig. 6) obtained from Eqs. (5)-(7) under the assumptions described above show that N_+ is characterized by a faster change with an increase in the Re concentration than N_-. Consequently, the quantity $R_+ \sim \sim 1/N_+$ must increase sharply, which is characteristic of a zone that was nearly filled [19]. Since the quantity N is [20] a complicated function of the conditions on the Fermi surface and it is proportional to m^*Bv_F where m^* is the effective mass and B is the area of the Fermi surface, the rapid change in N_+ indicates a considerable change in the hole sheet of the Fermi surface in this concentration region. The difference in the change in $|v_-|$ and $|v_+|$ (Fig. 7) is also characteristic of the behavior of carriers in a nearly empty and nearly full zone. Consequently, the changes in the Hall coefficient in the region of concentrations to 6 at.% Re are probably caused by changes in the zonal structure in accordance with the hard band model. This direction in the change of the Hall coefficient apparently must be preserved in the entire range of solid solutions. The maximum on the Hall /63 coefficient curve as a function of the concentration in the region 6-7 at.% Re caused by the change in the alloying effect on R shows that the value of the Hall coefficient is sensitive to changes taking place in the electron subsystem which are not related to changes in the parameters in the conductivity zone.

Since the positions of the anomalies on the volume resistivity and Hall coefficient curves as functions of the concentrations coincide (see Figs. 2, 3), it is probable that they have the same character and that they are due to the different
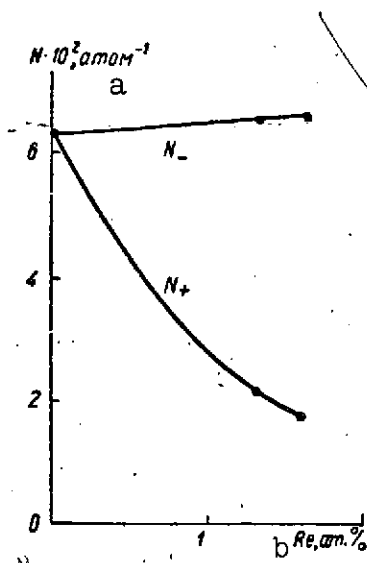
Fig. 6. Change in N_ and N+
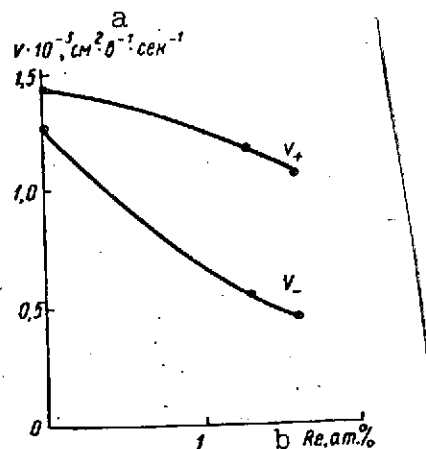as a result of alloying.

Key: a. Atom^{-1}
     b. Re, at.%

Fig. 7. Change in $|v_-|$ and $|v_+|$
as a result of alloying.

Key: a. $cm^2 \cdot V^{-1} \cdot sec^{-1}$
     b. Re, at.%

magnitudes of the scattering potential U_{Re} in concentration
regions lower than 6 and higher than 8 at.% Re.

Thus, even though the changes in the energy spectrum
in solid solutions correspond to the hard zone models, transfer
phenomena cannot be described within the framework of this
approximation. The determining factor for transfer phenomena
is probably the interaction of the carriers with the extrinsic
centers.

Angular Spectra of Annihilation Photons

As a result of the annihilation of the positron, two photons
are formed with the electron of the metal, whose angular distribu-
tion is described by the distribution of the pulses of the electron-
positron pairs. Since the life of the positron in the metal is
much longer than the thermolysis time, the magnitude of the
positron pulse compared with the electron pulse can be ignored,
and the angular distribution of the annihilation photons (ADAP)

will describe the pulse distribution of the electrons [21].
For an ideal degenerate Fermi gas, the ADAP has the form

$$I(\alpha) = C(\alpha_m^2 - \alpha^2),$$

(8)

where $\alpha = \pi - \phi$, ϕ is the angle of dispersion of the annihilation photons, $\alpha \simeq P_\perp/mC$, $P_\perp$ is the component of the electron pulse which is perpendicular to the emission direction of the photons, α_m is the maximum angle corresponding to the maximum pulse and C is a constant.

In the general case, the ADAP is a superposition of the parabolic component (8) and the Gaussian component

$$I(\alpha) = De^{-\frac{\alpha^2}{M^2}},$$

(9)

resulting from the annihilation of positrons with electrons localized around ionic cores [22].

The parameter M in (9) is related to the distance r_m from the core at which the product of the wave functions of the positron and electron have a maximum [22]:

$$M^2 = \frac{3 \cdot 10^6 \hbar}{m^2 c^2 r_m^2}.$$

(10)

Here m is the electron mass and c is the velocity of light. In transition metals, the Gaussian component part in the ADAP is anomalously large. This is due to the fact that the state of the valent electrons in metals with d-shells that were not filled is transient, from highly bonded electrons to nearly free electrons.

The measurements of the ADAP of tungsten and its alloys with Re were carried out using the method described in article [23]. Since the positrons annihilate with low electrons, not only with the electrons on the Fermi surface, this method is more sensitive to the admixtures than the transfer phenomena.

Therefore the alloys were smelted in an arc furnace with the consumable electrode. The ingots were subjected to multiple deformation with intermediate annealing to remove concentration <u>/64</u> inhomogeneities. The samples for the measurements were annealed for full recrystallization at 1800°C for a period of 2 hours in vacuum. All curves were reduced to the maximum intensity of 1000 pulses. To separate the two contributing components in (8) and (9), in the ADAP, the graphical analysis described in article [24] was carried out which gives reliable results when one of the components is dominant. The treatment of the ADAP obtained has shown that for tungsten and all alloys except W + 1.8% Re for angles $\alpha \geq 1 \cdot 10^{-3}$ rad it is described by relation (9) (Fig. 8). For tungsten $r_m = 0.87 \cdot 10^{-3}$ cm, which is closer to the value of the ionic radius (about $0.7 \cdot 10^{-3}$ cm) than the metallic radius ($1.4 \cdot 10^{-3}$ cm) [25].
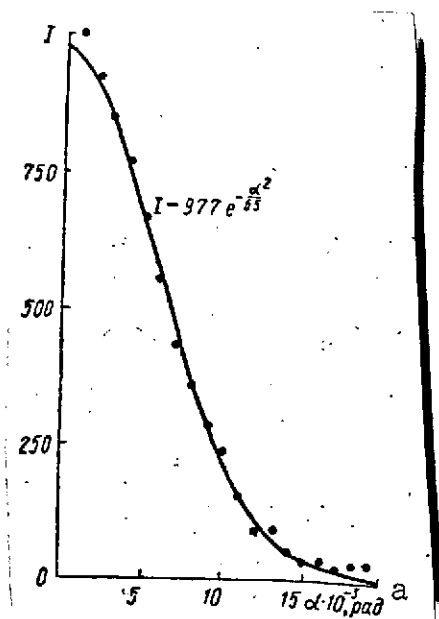
The values $\Delta r_m = r_{m\ all} - r_{m\ w}$ which were calculated from the values of M in the range to 5-6 at.% Re were negative and increase in absolute value with an increase in the Re concentration. Above 6 at.% Re, apparently Δr_m no longer increases, at 12 at.% Re, the values of Δr_m are close to zero and vary little all the way to the limiting concentrations (Fig. 9).

Since the wave function of the positron is the same for all alloys, the change in r_m reflects the change in the electron structure of tungsten during the alloying process. From the form of the curve
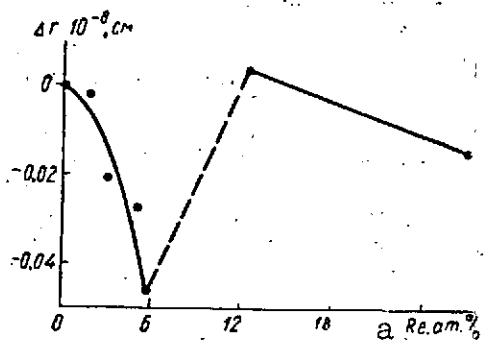

Fig. 8. Angular spectrum of photons during annihilation of positrons in tungsten.

Key: a. Radians

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Fig. 9. $\Delta r_m = r_{m\ all} - r_{m\ w}$ vs. concentration.

Key: a. Re, at.%

for Δr_m which depends on the concentration (Fig. 9), it follows that the electron structure of alloys does not change monotonically with an increase in the Re content, and that these changes do not occur in the conductivity zone, but characterize the degree of localization of the valent electrons. Since the parameter r_m reflects some effective dimension of the atom in the lattice, the different character of the effect of Re on r_m in concentration ranges below and above 6 at.% Re can probably explain the different signs of the deviations from the Vegard law for the crystal lattice period of solid W-Re /65 solutions in these concentration ranges (Fig. 10). To eliminate the possible effect of the fine structure, the period was measured when the Re content was equal to or larger than 6 at.% in EZMWC alloys.
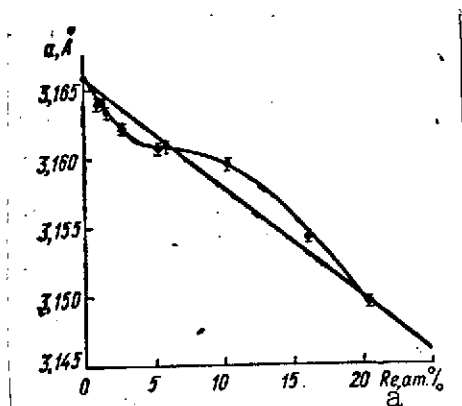

Fig. 10. Crystal lattice period of solid tungsten-rhenium solutions vs. concentration.

Key: a. Re, at.%

Magnetic Susceptibility

The magnetic susceptibility was measured at room temperature on apparatus similar to that described in article [26]. The samples for the measurements were cut out from EZMWC alloys.

The magnetic susceptibility in W-Re alloys increases with an increase in the Re concentration, for 6-7 at.%, the increment reaches values on the order of 15% compared

to pure tungsten. In regions where the concentrations are higher, the susceptibility does not change for all practical purposes (Fig. 11a).
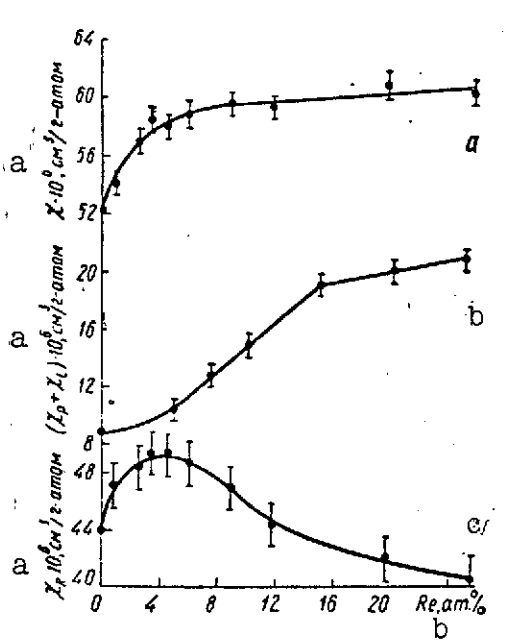

Fig. 11. Magnetic suscepti-
bility and its components
vs. concentration.

Key: a. Cm3/g·atom
     b. Re, at.%

The total measured suscepti-
bility χ can be represented as

$$\chi = \chi_P + \chi_L + \chi_{ion} + \chi_R.$$

where χ_P is the Pauli paramagnetism, χ_L is the Landau diamagnetism, χ_{ion} is the diamagnetism of the ionic cores, and χ_R is the residual para-magnetism.

The Pauli paramagnetism is determined by the density of the states on the Fermi surface

$$\chi_P = \mu_0^2 N (E_F).$$

(11)

where $\mu_0 = (g/2)\mu_B$, g is the spectroscopic splitting factor, μ_B is the Bohr magneton, which can be calculated from the data on the electron heat capacity: $C = \gamma T$ where $\gamma = (\pi^2/3)k^2 N(E_F)$. The Landau diamagnetism is equal to one third of the Pauli paramagnetism. In transition metals and alloys χ is ignored and it is assumed that m* is the effective mass which is much larger than the mass m of the electron. For tungsten, this assumption is not valid, since in tungsten the mean value m*/m is apparently close to 1 [9]. Therefore, for tungsten and its alloys with Re the total contribution of the conductivity electrons was taken as $(2/3)\mu_0^2 N(E_F)$.

The form of the dependence on the concentration of

$$\chi_P + \chi_L = \frac{2}{3}\mu_0^2 N(E_F).$$

calculated from the results of the measurements of C = γT [17] differs markedly from the dependence of χ on the concentration (Fig. 11). Like γ, the contribution due to the conductivity electrons increases smoothly to 15 at.% Re, and during the transition from pure tungsten to the W + 5 at.% Re alloy, the quantity $\chi_P + \chi_L$ increases only by $1.7 \cdot 10^{-6}$ cm^3/mole. The quantity $\chi - (2/3)\chi_P = \chi_{ion} + \chi_R$ increases as a result of this /66 at concentrations equal to or lower than 5 at.% Re and then decreases smoothly (Fig. 11c). Linear interpolation between the calculated values of $\chi_P + \chi_L$ was used for separation.

The diamagnetism of the filled shells is not taken into account uniquely; however, calculations carried out for the same approximations have shown that during the transition from tungsten to rhenium,χ_{ion} varies little [27]. Therefore, the form of the curve for $\chi_{ion} + \chi_R$ as a function of the concentration (Fig. 11c) reflects mainly the character of the change in χ_R. In articles [28-30] it was shown that in metals with d-shells that were not filled ,the term χ_R results from the contribution of the orbital magnetic moment through second-order perturbations to the paramagnetic susceptibility. This polarizing effect arises when the electric field containing the ion or atom does not have spherical symmetry. The magnitude χ_R in the strong bonding approximation is described by the expression

$$\chi_R = 4N\mu_B^2 \sum_{ij} f_i(1-f_j)\frac{1}{E_i - E_j}|<k,j|\,l_z\,|k,i>|^2. \tag{12}$$

where i, j are the indices of the bands, N is the Avogadro number, and the averaging is carried out over all k [30].

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Hence the orbital susceptibility is mainly determined by the energy width of the d-zone and the degree with which it is filled. Since detailed calculations are not available for tungsten, it is not clear in which direction χ_R will change only as a result of the filling of the tungsten conductivity band without changing its form, and to what extent the measurements of χ_R that were found in the Re alloying process correspond or do not correspond to the model of the hard zone. It can only be assumed that since the width of the tungsten conductivity band and the degree to which it is filled vary little in the solid solutions under consideration (which follows from the estimates obtained for the hard band model [15]), the changes in χ_R found in W-Re solid solutions are apparently caused by a change in the magnitude of the matrix element in (12), which in turn is determined by the wave functions of the d-electrons. Thus, on the basis of the curve for χ_R as a function of the concentration, the conclusion can be made that the character of the effect of Re atoms in the W lattice on the three-dimensional distribution of the electrons of the d-shell is different for Re concentrations above and below 5 at.%.

Conclusion

From the analysis of the properties of W-Re solid solutions which depend on the concentrations that was carried out above it follows that discontinuity in the constitutional diagram, a property found in the range of the same concentrations in all diagrams, can only be explained by the nonmonotonic effect of Re on the character of the distribution of the valent electrons around the points of the crystal lattice. This general conclusion can be made concrete by comparing the data from various methods.

The increment $\Delta\rho(x)$ in the volume electrical resistivity caused by the alloying is described well by the Nordheim rule in

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concentration ranges lower than or equal to 6 at.% Re and higher than 8 at.% Re. The relation $\Delta\rho(x) = Ax(1 - x)$ was obtained on the assumption that the effect of the admixture only manifests itself in the local change of the lattice potential [7]. Therefore, the different values of A for various concentration ranges indicate that the magnitude of these local perturbations varies in a relatively narrow concentration region. Since the value of A is determined by the degree by which the potential of the admixture differs from the potential of the unperturbed lattice, the magnitude of the scattering potential of the Re atom (the character of the distribution of its valent electrons) is different in different concentration regions but remains constant within each region. For the ADAP, the magnetic susceptibility, and probably the parameter of the crystal lattice, this effect manifests itself along the entire lattice in an averaged form. In accordance with the interpretation of the results obtained by the method of annihilation of electron-positron pairs, the change in the potential of the Re atom is related to the reduced degree of electron localization.

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